Journal of Food Engineering 147 (2015) 111–119

Contents lists available at ScienceDirect

Journal of Food Engineering

journal homepage: www.elsevier.com/locate/jfoodeng

Estimation of the effective diffusion coefficient of water in skim milk during single-drop drying

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ARTICLE INFO

Article history:
Received 7 August 2014
Received in revised form 16 September 2014
Accepted 21 September 2014
Available online 30 September 2014

Keywords:
Diffusion coefficient
Skim milk
Spray drying
Single drop
NMR
Parameter estimation

ABSTRACT

This paper presents a new approach combining experimental methodology and modelling, developed to evaluate the effective diffusivity of water in skim milk during drying over a full range of water contents and temperatures. This parameter is important to support modelling of spray-drying processes and designing of equipment. The effective diffusion coefficient is evaluated using a combination of nuclear magnetic resonance (NMR) and parameter estimation. NMR is used to determine the temperature dependence and parameter estimation is used to estimate the water concentration dependence of the effective diffusivity of water in skim milk (0.90 on total weight basis) during drying by comparing the experimental data obtained using a suspended-drop method, which allows the recording of weight and temperature changes during drying, with the results of a distributed heat and mass transport model. The results indicate that the free-volume theory best predicts the dependence of the effective diffusion coefficient of water in skim milk. A mathematical correlation of effective diffusivity over a full range of water contents and temperatures (from 50 to 90 °C) was obtained and experimentally successfully validated for concentrated skim milk (0.70 on total weight basis).

1. Introduction

Spray drying is the most common method for producing powder from a liquid feed. The liquid feed is sprayed at the top of a drying chamber into a flow of hot air. Spray drying involves numerous phenomena such as feed atomization, airflow, inter- and intraparticle heat and mass transfer, and particle interactions. These phenomena make the design and scale-up of the spray dryer an arduous task. However, modelling and simulating these phenomena support the design of spray dryers.

Drying an atomized feed involves simultaneous heat and mass transfer under the unsteady state of a spray consisting of single drops considered as spheres having their sizes in the range of 10–500 μm, depending on the product and application. The literature uses various drying models to describe the drying of a single drop during spray drying, and these can be classified, depending on the modes of heat and mass transfer, as follows:

- a lumped-parameter model for heat and mass transport (Chen, 2008; Chen and Xie, 1997; Langrish and Kockel, 2006);
- a lumped-parameter model for mass transfer and a distributed-parameter model for heat transfer (Farid, 2003; Mezhericher et al., 2008);
- a distributed-parameter model for mass transfer and a lumped-parameter model for heat transfer (Adhikari et al., 2007; Ferrari et al., 1989; Sano and Keey, 1982); and
- distributed-parameter models for heat and mass transport (Dalmaz et al., 2007; Shabde et al., 2005).

To identify which model is suitable for a given application, the dimensionless Biot numbers for heat ($Bi = (h_{ext}L_c)/k$) and mass ($Bi_m = (k_{ext}L_c)/D$) transport are used to compare the internal and external transport resistances. In the case of heat transport, external convection is compared with internal conduction, and for mass transport, external convection is compared with internal diffusion. Biot numbers for heat and mass transport lower than 0.1 indicate that the drying process is externally controlled and that a lumped-parameter model can be used to model it with an error typically 5% compared with the distributed model (Welty et al., 2009). Biot numbers greater than 0.1 indicate an internally controlled process for heat and mass transport and that a distributed-parameter model should be used. Biot numbers for mass transport during spray drying are usually >0.1 and, consequently

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http://dx.doi.org/10.1016/j.jfoodeng.2014.09.032
0260-8774/

Nomenclature

Latin alphabet

a_w	water activity (–)
Bi	Biot number for heat transport (–)
Bi_m	Biot number for mass transport (–)
C_A	number concentration (m^{-3})
c_{ext}	external vapour concentration ($kg\ m^{-3}$)
c_{sat}	vapour concentration on the surface ($kg\ m^{-3}$)
c_v	volume fraction ($m^3\ m^{-3}$)
C_p	specific heat ($J\ kg^{-1}\ K^{-1}$)
d	pure density ($kg\ m^{-3}$)
D	diffusion coefficient ($m^2\ s^{-1}$)
$D_{0,I}$	optimized parameter in Model I ($m^2\ s^{-1}$)
$D_{0,II}$	optimized parameter in Model II ($m^2\ s^{-1}$)
$D_{0,III}$	optimized parameter in Model III ($m^2\ s^{-1}$)
F	flux of water leaving the surface ($kg\ m^{-2}\ s^{-1}$)
h_{ext}	external heat transfer coefficient ($W\ m^{-2}\ K^{-1}$)
k	thermal conductivity ($W\ m^{-1}\ K^{-1}$)
k_{ext}	external mass transfer coefficient ($m\ s^{-1}$)
L_c	characteristic length (m)
M	molecular weight of water ($g\ mol^{-1}$)
n	mass flux ($kg\ m^{-2}\ s^{-1}$)
P_{sat}	vapour pressure (Pa)
r	radius coordinate (m)
R	radius of the drop (m)
R^2	coefficient of determination (–)
$\underline{R}$	ideal gas constant ($J\ K^{-1}\ mol^{-1}$)
t	time (s)
T	temperature (K)
RSS	residual sum of squares (–)
u	water mass fraction on a solid weight basis ($kg\ kg^{-1}$)
V	volume (m^3)
V_{VSOL}	water free volume in the solid solution (m^3)
v_s	velocity of a plane through which no net transport of the reference component occurs ($m\ s^{-1}$)
w	water mass fraction on a total weight basis ($kg\ kg^{-1}$)
z	solid fixed coordinate (kg)
ΔH_{ev}	latent heat of water evaporation ($J\ kg^{-1}$)

Greek alphabet

α	proportionality factor in Model III (–)
ρ	density ($kg\ m^{-3}$)

Subscripts

$_0$	initial value
$_A$	component A
$_B$	component B
$_b$	bound
$_{eff}$	effective
$_{ext}$	external
$_{exp}$	experimental
$_{max}$	maximum value
$_s$	solid
$_{sim}$	simulated
$_w$	water

drying is fully governed by internal diffusion in the drops (van der Lijn, 1976).

In the case of spray drying of a complex product such as milk, the distributed heat transport model of spray drying has been widely published and suitable parameter values for the model (e.g. k, C_p, and h_{ext}) can be found in the literature (Dalmaz et al., 2007; Sano and Keey, 1982). This is not the case for mass transport in spray drying, for which few fundamental models of moisture transport during spray drying have been published (Chen and Xie, 1997; Ferrari et al., 1989). The moisture loss during spray drying creates moisture gradients within the particles, while diffusion mechanisms such as molecular diffusion, capillary flow, Knudsen diffusion, hydrodynamic flow, and surface diffusion control the mass transport. It is generally accepted that diffusion mechanisms in spray drying are dependent on the temperature and water content and affected by structural changes, but a full understanding of the mechanisms involved and their importance for moisture loss in spray drying have not been completely clarified.

To overcome the lack of understanding of these matters, a modified Fick's second law describing mass transport in terms of effective diffusivity has been proposed as a suitable model for designing equipment (Mujumdar, 2007). Effective diffusivity, originally formulated to describe mass transport in porous media (Bird et al., 2006; Cussler, 1997), depends on the water concentration, temperature, porosity and tortuosity of the medium used. Experimental evaluation of the effective diffusion coefficient as a function of water concentration and temperature is difficult, as no standard methods are available and the data reported in the literature are based on comparing the results of diffusion experiments with the solution of the diffusion equation for particular cases. Regular regime (RR) theory is an example of a method used to calculate the concentration and temperature dependence of the effective diffusion coefficient of water in skim milk (Ferrari et al., 1989). Ferrari et al. (1989) applied RR theory to a single gelled drop of skim milk with an initial diameter of 10 mm; they suggested a correlation to evaluate the effective diffusion coefficient as a function of water content and temperature, a correlation validated for water contents of 0.25–0.45 on a total weight basis and temperatures of 30–70 °C. Recently, Perdana et al. (2014) adapted this method to evaluate the mutual diffusivity of water in sucrose, lactose, and maltodextrin in a thin slab over a greater range of water contents, demonstrating the possibility of using this method for complex solutions. One limitation of this method is that it allows the estimation of effective diffusivity in only a limited range of water contents below 0.65 on a total weight basis.

A standard method does not exist either to experimentally measure or mathematically describe the effective diffusion coefficient.

As an alternative to the RR method, the effective diffusivity can be estimated using parameter estimation techniques, i.e. by optimizing the diffusivity value that minimizes the difference between the fitting of the experimental drying kinetics data to the suitable mathematical models describing the experiments in terms of heat and mass transport. Because these models are complex, non-linear, and collinearity may exist between parameters, a suitable experimental and modelling approach is necessary to guarantee the accuracy of the diffusivity values estimated and the use of a larger range of water content.

The aim of this work is to present a new approach combining experimental methodology and modelling to evaluate the effective diffusivity of water in skim milk over a full range of water contents and temperatures.

2. Experimental

2.1. Material

Drying experiments and NMR measurements were performed on skim milk of different initial concentrations. Fresh milk was